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**NONPROVISIONAL PATENT  
APPLICATION TRANSMITTAL RULE §1.53(b)  
IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

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Commissioner for Patents  
Washington, D.C. 20231

Sir:

Transmitted herewith for filing under 37 C.F.R. §1.53(b) is a nonprovisional patent application:

For (Title): FERROMAGNETIC ZnO-TYPE COMPOUND INCLUDING TRANSITION  
METALLIC ELEMENT AND METHOD FOR ADJUSTING  
FERROMAGNETIC CHARACTERISTICS THEREOF

By (Inventors): Hiroshi YOSHIDA; Kazunori SATO

- ☒ 25 pages of Specification is attached.  
☒ Drawings (4 sheets) is attached.  
☒ A Declaration and Power of Attorney is attached.  
☒ An assignment of the invention to ROHM CO., LTD. is attached, along with  
 Form PTO-1595 and a check for \$40.00.  
☒ A certified copy of Japanese application(s) No.(s) 11-308911; dated October 29, 1999

The filing fee is calculated below and includes claim status after entry of any Preliminary Amendment noted above:

			SMALL ENTITY			LARGE ENTITY	
FOR:	NO. FILED	NO. EXTRA	RATE	FEE	OR	RATE	FEE
BASIC FEE				\$ 355	OR		\$ 710
TOTAL CLAIMS	10 - 20	= 0	x 9 =		OR	x 18	\$ 0
INDEP CLAIMS	3 - 3	= 0	x 40 =		OR	x 80	\$ 0
<input type="checkbox"/> MULTIPLE DEPENDENT CLAIMS			+135 =		OR	+270	\$ 0
			TOTAL		OR	TOTAL	\$ 710

- ☒ A Check No. 303626 in the amount of \$750.00 is attached. Please charge any fee deficiency or credit any overpayment to Deposit Account No. 01-2300.

Respectfully submitted,

George E. Oram, Jr.  
Registration No. 27,931

GEO/hk

S P E C I F I C A T I O N

BE IT KNOWN THAT We, HIROSHI YOSHIDA residing at 82-4, Daiwahigashi 2-chome, Kawanishi-shi, Hyogo, Japan and KAZUNORI SATO residing at Daiichihukuwaso A-12, 2-36, Makiochi 5-chome, Minoh-shi, Osaka, Japan, subjects of Japan, have invented certain new and useful improvements in

FERROMAGNETIC ZnO-TYPE COMPOUND INCLUDING TRANSITION  
METALLIC ELEMENT AND METHOD FOR ADJUSTING  
FERROMAGNETIC CHARACTERISTICS THEREOF

of which the following is a specification:-

## 5

The present invention relates to a single-crystalline ferromagnetic ZnO-type compound realizing ferromagnetic characteristics by crystal-mixing at least one of transition elements such as V, Cr, Fe, Co, Ni, Rh or Ru to a light-transmitting ZnO-type compound and a method for adjusting ferromagnetic characteristics thereof. More particularly, it relates to a ferromagnetic ZnO-type compound exhibiting desired ferromagnetic characteristics, such as ferromagnetic transition temperature, and a method for adjusting the ferromagnetic characteristics thereof.

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If it is possible to obtain single-crystalline ferromagnetic thin films exhibiting high ferromagnetic characteristics while transmitting light, it will be possible to provide light isolators or to perform high-density magnetic memory as it is required in transmitting a large amount of information and thus to manufacture electronic materials required for transmission of large amount of information in the future. It is therefore being wanted for materials exhibiting

ferromagnetic characteristics while transmitting light.

On the other hand, ZnO-type compounds are advantaged through their characteristics of exhibiting a large band gap of 3.3 eV and transmitting light of wavelengths ranging from blue to ultraviolet, and due to their large binding energy of their excitons when compared to, for instance, GaN, remarkable progresses are being expected in the field of manufacturing optical devices such as light quantum computers employing coherent spin states once it is possible to exhibit ferromagnetism through these materials. However, while there are conventionally known cases in which ZnO is doped using Mn, ZnO will be in an antiferromagnetic state and no cases have so far been reported in which ZnO-type compounds are in ferromagnetic conditions.

As noted, if it would be possible exhibit ferromagnetic characteristics by using ZnO-type compounds, quite a large variety of applications to devices utilizing magnetic-optical effects will be provided in which these compounds are used in combination with light-emitting elements such as semiconductor laser of ZnO-type compounds exhibiting large binding energy of excitons or in which light reflecting magnetic states are generated.

In case of arranging memories of ferromagnetic bodies by irradiating such light and varying magnetic states, it will be necessary to set ferromagnetic

characteristics to be desired ones in which, for instance,  
a ferromagnetic transition temperature (Curie  
temperature) is set to be a temperature that is varied upon  
radiation of light (a temperature slightly higher than  
5 room temperature).

#### SUMMARY OF THE INVENTION

The invention has been made in view of these  
circumstances, and it is an object thereof to provide a  
10 ferromagnetic ZnO-type compound exhibiting ferromagnetic  
characteristics by using a light-transmitting ZnO-type  
compound.

It is another object of the present invention to  
provide a method for adjusting ferromagnetic  
15 characteristics of the ferromagnetic ZnO-type compound in  
obtaining the ferromagnetic ZnO-type compound which  
ferromagnetic characteristics such as ferromagnetic  
transition temperatures may be adjusted.

The inventors of the present invention have found  
20 upon performing various studies for obtaining a  
single-crystalline structure with ferromagnetic  
characteristics by using a ZnO-type compound that is a  
particularly suitable material transmitting light, that  
the ion radii of transition elements such as Ti, V, Cr,  
25 Mn, Fe, Co or Ni approximate to that of Zn and that  
single-crystalline structures may be sufficiently  
obtained even by substituting (crystal-mixing) more than

50% of Zn, that even though only antiferromagnetic characteristics may be obtained by crystal-mixing Mn to ZnO, ferromagnetic characteristics may be obtained adding holes or electrons (increasing or decreasing electrons) to the electron condition of Mn in this state (with 5 d-electrons), that similar effects as by adding holes to Mn may be achieved by crystal-mixing Cr or V, which number of d-electrons are less than those of Mn, to the ZnO-type compound, and that similar effects as by doping electrons to Mn may be achieved by crystal-mixing Fe, Co or Ni to the ZnO-type compound, and finally came to find out that by crystal-mixing at least one of transition elements such as V, Cr, Fe, Co, Ni, Rh or Ru to the ZnO-type compound, a stable ferromagnetic condition may be achieved by simply crystal-mixing these metallic elements.

By further continuing these studies, the inventors have found that transition elements such as Ti, V, Cr, Mn, Fe, Co or Ni assume high spin states with electron spins of  $5/2$ ,  $4/2$ ,  $3/2$ ,  $2/2$  or  $1/2$ , that ferromagnetic transition temperatures are variable by varying their densities, performing crystal-mixing by varying combinations or ratios of more than two types thereof or by adding n-type and/or p-type dopants, that the ferromagnetic state may be stabilized than compared to antiferromagnetic or paramagnetic states, that an energy in the ferromagnetic state (for instance, an energy with which the ferromagnetic state may be maintained in an

ordinary condition though an antiferromagnetic state is assumed in the presence of slightest differences) is adjustable, and that desired filtering functions may be exhibited by selectively performing crystal-mixing of more than two types of the above transition metallic elements that exhibit different minimum transmission wavelengths, and finally came to find out that by adjusting densities or mixing ratios of these transition metallic elements, a single-crystalline and ferromagnetic ZnO-type compound exhibiting desired magnetic characteristics may be obtained.

The ferromagnetic ZnO-type compound according to the present invention is a ZnO-type compound containing at least one metal selected from a group of transition metallic elements consisting of V, Cr, Fe, Co, Ni, Rh and Ru.

It should be noted that the term "ZnO-type compound" here denotes an oxide including Zn, a concrete example of which is ZnO, respective oxides of a IIA group element with Zn, a IIB group element with Zn or IIA group element and IIB group element with Zn.

With this structure, with the above-listed transition metallic elements which iron radii approximate to those of II group elements such as Zn, single-crystalline structures of Wurtzite structure may be maintained even upon substituting not less than 50 at% of Zn, and will exhibit ferromagnetic characteristics with

increased holes or electrons than compared to Mn while maintaining its transparency.

By containing at least two types of metals selected from a group consisting of the above metals, Ti, Mn and Cu, conditions of d-electrons of the metallic elements will be respectively different so that ferromagnetic characteristics will be varied in a more direct manner than achieved by doping holes or electrons so that ferromagnetic characteristics such as ferromagnetic transition temperatures may be adjusted.

By performing doping by using at least either one of an n-type dopant or p-type dopant, the dopant will enter a matrix of ZnO so that it will act on d-electrons close to the matrix of ZnO, though their effects will not be so much direct as it is the case with transition element, and holes or electrons are varied to enable adjustments of ferromagnetic characteristics.

According to the method for adjusting ferromagnetic characteristics of a ZnO-type compound, at least one of

- (1) at least one metallic element selected from a group consisting of transition metallic elements of V, Cr, Fe, Co, Ni, Rh and Ru,
- (2) at least two metallic elements selected from a group consisting of the above transition metallic elements, Ti, Mn and Cu, and
- (3) at least one of the above (1), (2), an n-type dopant, and a p-type dopant



is added to the ZnO-type compound for adjusting ferromagnetic characteristics by adjusting densities of the above transition elements, Ti, Mn, Cu or n-type dopant or p-type dopant or by varying combinations of these  
5 metallic elements.

Particularly, it is possible to adjust the ferromagnetic transition temperature to be a desired temperature by using at least one method of adjusting the density (the density of transition elements, metallic  
10 elements such as Mn and dopants), and varying combinations of metallic elements as listed in above (2); to stabilize the ferromagnetic state by adjusting the ferromagnetic energy by crystal-mixing at least two types of metallic elements as listed in above (2) and by decreasing the  
15 entire energy through kinetic energy by holes or electrons introduced by the metallic elements themselves; and to further stabilize the ferromagnetic state by crystal-mixing at least two types of metallic elements as listed in above (2) and by controlling the size and sign of  
20 magnetic interaction between metallic atoms through holes or electrons introduced by the metallic elements themselves.

It is further possible to obtain a ferromagnetic ZnO-type compound with desired light-filtering  
25 characteristics by crystal-mixing at least two types of metallic elements as listed in above (2), by controlling the size and sign of magnetic interaction between metallic

atoms through holes or electrons introduced by the metallic elements themselves and by controlling transmitting characteristics of light owing to crystal-mixing of metallic elements.

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#### BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is an explanatory view showing one example of a apparatus for forming a ferromagnetic ZnO-type compound thin film according to the present invention;

10 Fig. 2 is a diagram illustrating differences  $\Delta E$  between entire energy of antiferromagnetic body and entire energy of ferromagnetic body when a transition element such as V, Cr is crystal-mixed to ZnO;

15 Figs. 3(a) and 3(b) are diagrams illustrating variations in ferromagnetic transition temperatures and magnetic moments upon variations in density of transition element that is crystal-mixed to ZnO;

20 Figs. 4(a) and 4(b) are diagrams illustrating variations in ferromagnetic transition temperatures depending on ratios of crystal-mixing more than two types of transition elements; and

Fig. 5 is an explanatory view showing variations in magnetic states when an n-type or p-type dopant is added to, for instance, Mn.

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#### DETAILED DESCRIPTION

The ferromagnetic ZnO-type compound and the method

for adjusting its ferromagnetic characteristics according to the present invention will now be explained with reference to the drawings. The ferromagnetic ZnO-type compound according to the present invention is  
5 a ZnO-type compound containing at least one metal selected from a group consisting of transition metallic elements of V, Cr, Fe, Co, Ni, Rh and Ru.

As already described, the inventors of the present invention have found out through various studies for  
10 obtaining a ferromagnetic material using a ZnO-type compound that in case of transition elementlic elements V, Cr, Fe, Co, Ni, Rh and Ru, these transition elementlic elements exhibit ferromagnetism by performing crystal-mixing of only these elements in a single manner due to  
15 the fact that 3d-electrons are increased/decreased through Mn, which itself is antiferromagnetic, as indicated by the difference  $\Delta E$  between antiferromagnetic entire energy and ferromagnetic entire energy as illustrated in Fig. 2. While the ratio of mixed crystal  
20 to Zn in ZnO is 25 at% in this example, ferromagnetism can also be achieved already with a mixed crystal ratio of several %, causing no damages on crystallinity and transparency though this ratio be increased, and satisfactory ferromagnetism can be achieved with a ratio  
25 of 1 at% to 99 at%, preferably 10 at% to 80 at%. The transition metallic element may be used singly or, as it will be described later, by mixed crystal (alloying) more

than two types thereof.

For forming a thin film using such a ZnO-type compound containing a transition metallic element, a substrate 5 that is made, for instance, of sapphire for growing a ZnO-type compound is disposed on a substrate holder 4 within a chamber 1 capable of maintaining an ultrahigh vacuum condition of approximately  $1.33 \times 10^{-6}$  Pa, wherein the substrate 5 may be heated through a heater 7 as illustrated in a schematic explanatory view of a MBE apparatus of Fig. 1. A cell 2a containing therein an elemental material (source) Zn comprising the compound to grow, a cell 2b containing therein a transition element such as Fe (though only one cell is illustrated, more than two cells are provided when crystal-mixing more than two types of metals), a cell 2c containing therein an n-type dopant such as Ga, Al or In, and RF radical cells 3a, 3b for generating radical oxygen O and radical nitrogen N are disposed to oppose the substrate 5 held by the substrate holder 4. It should be noted that solid materials such as Zn or transition metallic elements might assume atomic states by putting oxides of these metals into the cells.

Though not illustrated, the cells 2a to 2c into which the solid bodies (elements) are put are respectively provided with heaters such that solid body sources are put into atomic states for evaporation through heating, and the radical cells 3a, 3b are activated by RF (radio

frequency) coils 8 as shown in the drawing. As for the Zn, transition metallic elements and n-type dopant materials, a solid body source of a purity of 99.99999% is put into an atomic state while an atomic gas of 99.99999% in which O<sub>2</sub> is activated through the RF radical cells is used as O, and N<sup>+</sup> or N<sub>2</sub> in an excited state is used by activating N<sub>2</sub> molecules or N<sub>2</sub>O in the radical cells. It should be noted that Ga or transition metallic elements might also be put in an atomic state by irradiating electromagnetic waves of microwave regions to molecular gas.

While making ZnO grow, Ga serving as an n-type dopant, an atomic N as a p-type dopant, and an atomic transition element such as Fe are simultaneously poured onto the substrate 5 at a flow rate of  $1.33 \times 10^{-5}$  Pa, a flow rate of  $6.65 \times 10^{-5}$  Pa, and a flow rate of  $1.33 \times 10^{-5}$  Pa, respectively, for growing at a temperature of 350 to 750°C in order to grow a ZnO thin film 6 crystal-mixed with a transition metallic element. While doping is exemplarily performed using an n-type dopant or a p-type dopant in the above explanations, no dopants are used for the doping but doping is only performed using transition elements including Mn, Ti and Cu in the examples as illustrated in Fig. 2 as well as Tables 1 and 2 as will be described later.

The ZnO thin film in which V, Cr, Fe, Co and Ni are crystal-mixed exhibits ferromagnetism as shown in Fig. 2

in which V, Cr, Fe, Co and Ni respectively exhibit a difference  $\Delta E$  between antiferromagnetic energy and ferromagnetic energy of  $20 \times 13.6$  meV,  $15 \times 13.6$  meV,  $10 \times 13.6$  meV,  $14 \times 13.6$  meV and  $18 \times 13.6$  meV. It should  
5 be noted that the data of Fig. 2 are data obtained through first principle calculation in which simulation is performed using atomic numerals as input parameters.

While the ZnO compound is doped with a transition  
elementlic element in this example, similar structures as  
10 ZnO may be obtained also with ZnO-type compounds in which a part of Zn of ZnO is substituted by other II-group elements such as Mg or Cd, and it is similarly possible to obtain a ferromagnetic single-crystalline structure.

According to the ferromagnetic ZnO-type compound of  
15 the present invention, a transition metallic element having ion radius that are substantially identical to that of Zn are crystal-mixed such that  $Zn^{2+}$  is substituted by  $Fe^{2+}$  or similar of the transition metallic element for maintaining the Wurtzite structure. Moreover, since the  
20 above transition metallic elements such as Fe assume electron structures in which d-electrons are increased than compared to Mn, the ferromagnetic state is stabilized as it is as illustrated in Fig. 2. Further, as illustrated in Tables 1 and 2 as will be described later, this  
25 ferromagnetic ZnO exhibits a large magnetic moment so that a Fe-containing ZnO-type compound having a larger magnetic moment  $4.04 \times 9.274$  J/T ( $4.04 \mu_B$  (Bohr magneton)) than that

of a Fe element (magnetic moment  $2 \times 9.274 \text{ J/T}$  ( $2 \mu_B$ )), and it is possible to obtain a ferromagnetic magnet with extremely strong magnetism.

Variations in magnetic characteristics have then  
5 been tested by varying densities of transition metallic  
elements. In addition to the above examples in which  
transition elementlic metals of a density of 25 at% are  
contained, those of a density of 50 at% were manufactured  
for testing respective magnetic moments ( $\times 9.274 \text{ J/T}$ ) and  
10 ferromagnetic transition temperatures (degree K). The  
magnetic moments and ferromagnetic transition  
temperatures have been obtained by measuring magnetizing  
rates using a SQUID (super conducting quantum interference  
device). The results are shown in Tables 1 and 2. It is  
15 obvious from Tables 1 and 2 that the ferromagnetic  
transition temperature tends to rise the higher the  
crystal-mixing ratio becomes (the higher the density  
becomes) and increases substantially proportional to the  
crystal-mixing ratio. This relationship is shown in Fig.  
20 3. It can further be understood that the ferromagnetic  
interaction between spins is also increased in accordance  
with increased in densities of transition elementlic  
elements and that the magnetic moments increase.

Table 1: Magnetic characteristics with transition elements of 25 at%

Type of transition element	Density of transition element (at%)	Magnetic moment ( $\mu_B$ )	Ferromagnetic transition temperature (degree K)
V	25	2.42	451
Cr	25	3.80	600
Fe	25	4.04	786
Co	25	2.96	528
Ni	25	1.88	389

Table 2: Magnetic characteristics with transition elements of 50 at%

Type of transition element	Density of transition element (at%)	Magnetic moment ( $\mu_B$ )	Ferromagnetic transition temperature (degree K)
V	50	2.58	681
Cr	50	3.95	792
Fe	50	4.02	959
Co	50	2.97	765
Ni	50	1.89	698

As already described, the transition metallic elements will assume high spin states with electron spins  $s$  of  $5/2$ ,  $4/2$ ,  $3/2$ ,  $2/2$ ,  $1/2$ , and as it is further evident from the Tables 1 and 2 as well as Figs. 3(a) and 3(b), it is possible to adjust and control the ferromagnetic interaction between spins and the ferromagnetic temperatures by varying the densities. It should be noted that it is preferable in view of practical use that the ferromagnetic transition temperature be not less than 150 degree K.

The inventors of the present invention have further



found out that it is possible to adjust conditions of holes or electrons and to concurrently exhibit respective magnetic characteristics by crystal-mixing more than two types of transition metallic elements. For example, Fe and Mn were crystal-mixed to be 25 at% as a whole and values  $x$  of  $\text{Fe}_{0.25-x}\text{Mn}_x\text{Zn}_{0.75}\text{O}$  were respectively varied. It was consequently possible to largely vary the ferromagnetic transition temperature, to be 0 degree K when  $x=0.15$  is satisfied, and to select a range for  $x$  between 0 to 0.15 for setting a desired ferromagnetic transition temperature as illustrated in Fig. 4(a). By similarly crystal-mixing Fe and Co to be 25 at% and respectively varying values  $x$  of  $\text{Fe}_{0.25-x}\text{Co}_x\text{Zn}_{0.75}\text{O}$ , ferromagnetic transition temperatures may be varied while maintaining a ferromagnetic state as illustrated in Fig. 4(b). Though not shown in the drawings, it is also possible to obtain magnetic moments in accordance with respective mixing ratios.

While ferromagnetic characteristics were varied by doping not less than two types of transition metallic elements in the above examples, the amount of holes or electrons may similarly be varied by doping an n-type dopant or a p-type dopant for varying the ferromagnetic states. In this case, the n-type or p-type dopant will enter a conduction band or a valence band of ZnO to act on adjoining d-electrons of transition metallic elements such that not all of the doped dopants will act as a whole,

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transition metallic elements, magnetic moments themselves will maintain constant values adjustments depending on transition elements materials that are crystal-mixed to ZnO in case of performing adjustments  
5 using dopants.

The n-type dopant may be B, Al, In, Ga, Zn or H, and raw materials for the doping may also be oxides of the above. The donor density is preferable not less than  $1 \times 10^{18} \text{ cm}^{-3}$ . In case doping is performed in a density of approximately  
10  $10^{20}$  to  $10^{21} \text{ cm}^{-3}$ , it will correspond to a crystal-mixing ratio of approximately 1 to 10%. The p-type dopant may be atomic like N, as explained above, such as  $\text{N}^+$  or  $\text{N}_2$  in an excited state. While it is hard to perform doping using a p-type dopant, the p-type density may set to be large  
15 by simultaneously doping a slight amount of n-dopant.

Upon performing further studies, the inventors have found out that minimum transmitted wavelengths differ depending on transition elementlic materials to be crystal-mixed to ZnO, that the minimum wavelength of  
20 transmitted light is adjustable by mixing not less than two types of transition metallic elements to be crystal-mixed, and that light filter for cutting light having a wavelength that is not more than a desired one may be formed. In other words, a ferromagnetic ZnO-type  
25 compound that transmits light of a desired wavelength may be obtained. The minimum wavelengths of light to be transmitted when crystal-mixing 25 at% of the respective

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Table 1. Demographic characteristics of the study population	
Age (years)	65.2 (SD 8.5)
Gender	
Male	55.2%
Female	44.8%
Education (years)	12.5 (SD 2.1)
Marital status	
Married	68.5%
Widowed	22.1%
Divorced	8.3%
Single	1.1%
Income (USD/month)	1,250 (SD 350)
Health status	
Good	72.3%
Fair	18.7%
Poor	9.0%

Type of transition element	Density of transition element (at%)	Minimum wavelength (nm)
ZnO:V	25	375
ZnO:Cr	25	426
ZnO:Fe	25	412
ZnO:Co	25	415
ZnO:Ni	25	390
ZnO:Mn	25	485

Table 1. Demographic characteristics of the study population	
Age (years)	65.2 (SD 8.5)
Gender	
Male	55.2%
Female	44.8%
Education (years)	12.5 (SD 2.1)
Marital status	
Married	68.5%
Widowed	22.1%
Divorced	8.3%
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Single	1.1%
Income (USD/month)	1,250 (SD 350)
Health status	
Good	72.3%
Fair	18.7%
Poor	9.0%

to perform film forming by using a MOCVD (metal organic  
chemical vapor deposition) apparatus. In this case,  
metallic materials such as Zn or transition element are  
introduced into the MOCVD apparatus in a form of an organic  
5 metallic compound such as dimethyl zinc. By using such MBE  
or MOCVD methods, it is possible to perform film forming  
in a non-equilibrium state and to dope transition metallic  
elements at desired densities. The film forming methods  
for the thin films are not limited to the above methods,  
10 and it is alternatively possible to form thin films through  
a laser abrasion method in which Zn oxide solid bodies,  
transition metallic element metals or solid bodies of  
oxides are used as targets and wherein film forming is  
performed while irradiating activated oxygen on the  
15 substrate.

In case of performing doping using transition  
metallic elements or oxides thereof as raw materials, it  
is also possible to use ECR plasma in which atomic states  
are assumed through electron excitation using radio  
20 frequency waves, laser, X rays or electron beams. ECR  
plasma may also be employed when using an n-type or p-  
type dopant. Such ECR dopant is advantaged in that doping  
may be performed in a atomic state up to a high density.

According to the present invention, a ferromagnetic  
25 single-crystalline structure may be obtained by simply  
making the ZnO-type compound contain transition metallic  
elements, application to high-performance communication

of information and quantum computers is enabled for use in quantum computers and large-capacity optical magnetic memory and as optical electronics materials covering a scope ranging from visible-light to ultraviolet regions by combining these with ZnO that are already realized and used as n-type and p-type transparent electrodes, or with optical fibers.

Although preferred example have been described in some detail it is to be understood that certain changes can be made by those skilled in the art without departing from the spirit and scope of the invention as defined by the appended claims.

What is claimed is:

1. A ferromagnetic ZnO-type compound, wherein a ZnO-type compound contains at least one metal selected from a group of transition metallic elements consisting  
5 of V, Cr, Fe, Co, Ni, Rh and Ru.

2. The ferromagnetic ZnO-type compound of claim 1, wherein said ZnO-type compound contains at least two types of metals selected from a group consisting of said transition metallic elements, Ti, Mn and Cu.

10 3. The ferromagnetic ZnO-type compound of claim 1, wherein said ZnO-type compound is doped at least either one of n-type dopant and p-type dopant.

4. A ferromagnetic ZnO-type compound, wherein a ZnO-type compound is added with at least one of  
15 (1) at least one metallic element selected from a group consisting of transition metallic elements of V, Cr, Fe, Co, Ni, Rh or Ru,  
(2) at least two metallic elements selected from a group consisting of said transition metallic elements, Ti, Mn  
20 and Cu, and  
(3) at least one of said (1), (2), an n-type dopant, and a p-type dopant

such that a desired ferromagnetic transition temperature is achieved.

25 5. A ferromagnetic ZnO-type compound in which addition of any one of (1) to (3) of claim 4 is performed to exhibit desired light-filtering characteristics.

6. A method for adjusting ferromagnetic characteristics of a ferromagnetic ZnO-type compound, wherein at least one of

(1) at least one metallic element selected from a group consisting of transition metallic elements of V, Cr, Fe, Co, Ni, Rh or Ru,

(2) at least two metallic elements selected from a group consisting of said transition metallic elements, Ti, Mn and Cu, and

(3) at least one of said (1), (2), an n-type dopant, and a p-type dopant

is added to said ZnO-type compound for adjusting ferromagnetic characteristics by adjusting densities of said transition metallic elements, Ti, Mn, Cu or n-type dopant or p-type dopant or by varying combinations of these metallic elements.

7. The adjusting method of claim 6, wherein the ferromagnetic transition temperature is adjusted to a desired temperature using at least one method by adjusting the density or by varying combinations of metallic elements as listed in said (2).

8. The adjusting method of claim 6, wherein the ferromagnetic state is stabilized by adjusting ferromagnetic energy by crystal-mixing at least two types of metallic elements as listed in said (2) and by decreasing the entire energy through kinetic energy by holes or electrons introduced by said metallic elements



themselves.

9. The adjusting method of claim 6, wherein the ferromagnetic state is stabilized by crystal-mixing at least two types of metallic elements as listed in said (2) and by controlling the size and sign of magnetic interaction between metallic atoms through holes or electrons introduced by said metallic elements themselves.

10. The adjusting method of claim 6, wherein a ferromagnetic ZnO-type compound with desired light-filtering characteristics is obtained by crystal-mixing at least two types of metallic elements as listed in said (2), by controlling the size and sign of magnetic interaction between metallic atoms through holes or electrons introduced by said metallic elements themselves and by controlling transmitting characteristics of light owing to crystal-mixing of said metallic elements.

## Abstract of the Disclosure

The ZnO-type compound contains at least one metal selected from a group of transition elements consisting of V, Cr, Fe, Co, Ni, Rh and Ru. Ferromagnetic characteristics are adjusted by adjusting densities of these transition elements, by varying combinations of more than two types of metals including Mn, or by adding dopants. It is consequently possible to obtain a ferromagnetic ZnO-type compound exhibiting ferromagnetic characteristics by using a light-transmitting ZnO-type compound, and to obtain a ferromagnetic ZnO-type compound exhibiting desired ferromagnetic characteristics.

FIG. 1

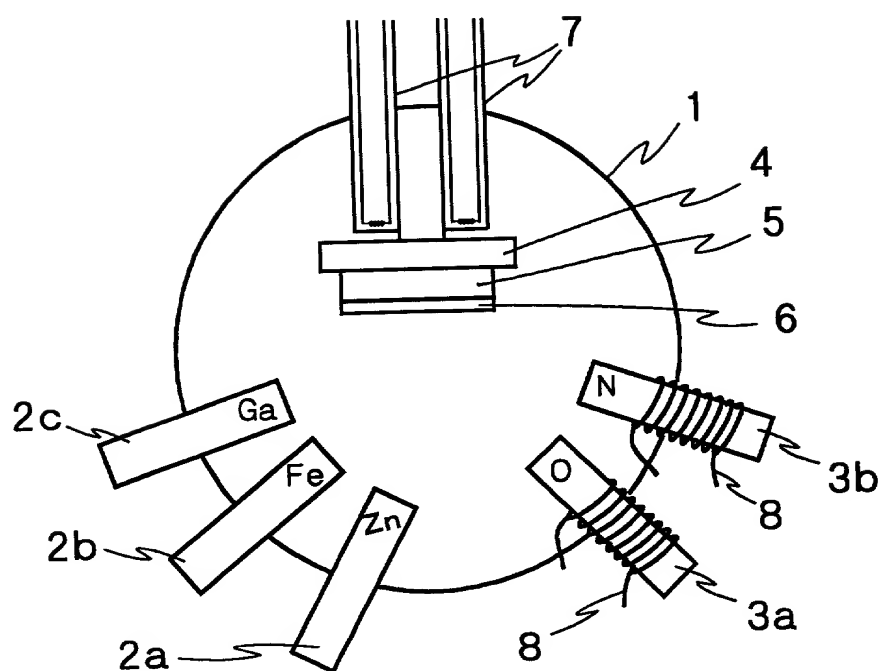


FIG. 2

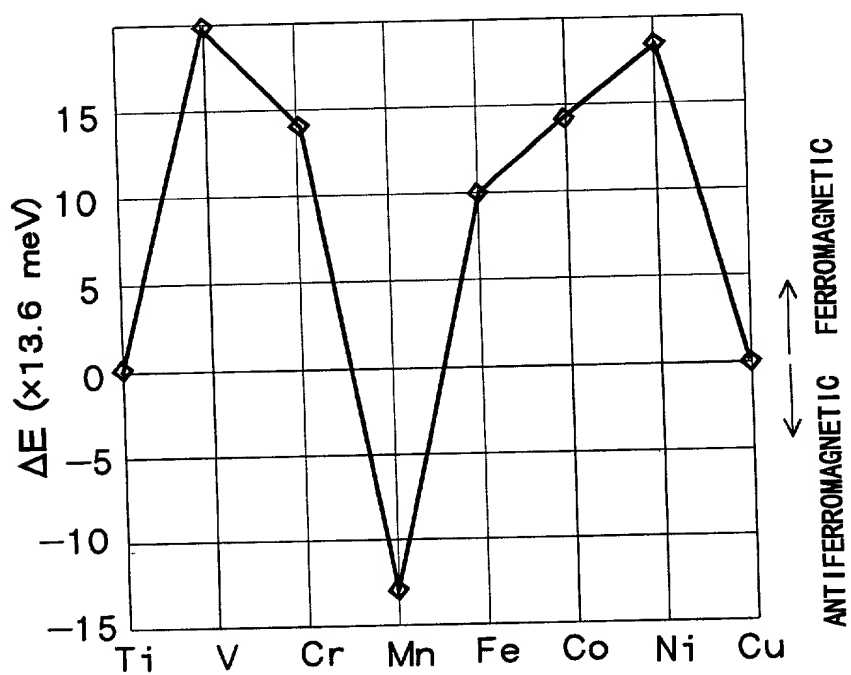


FIG. 3 (a)

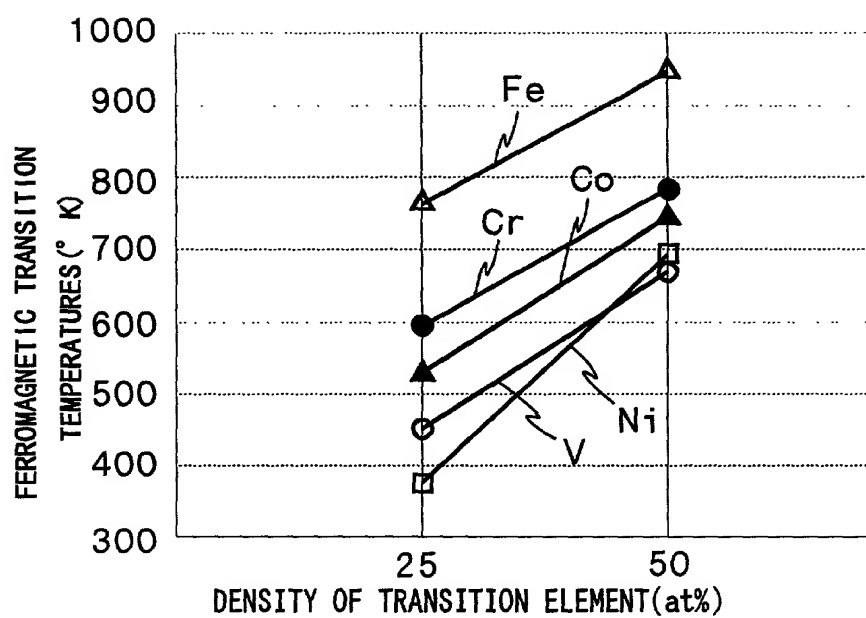


FIG. 3 (b)

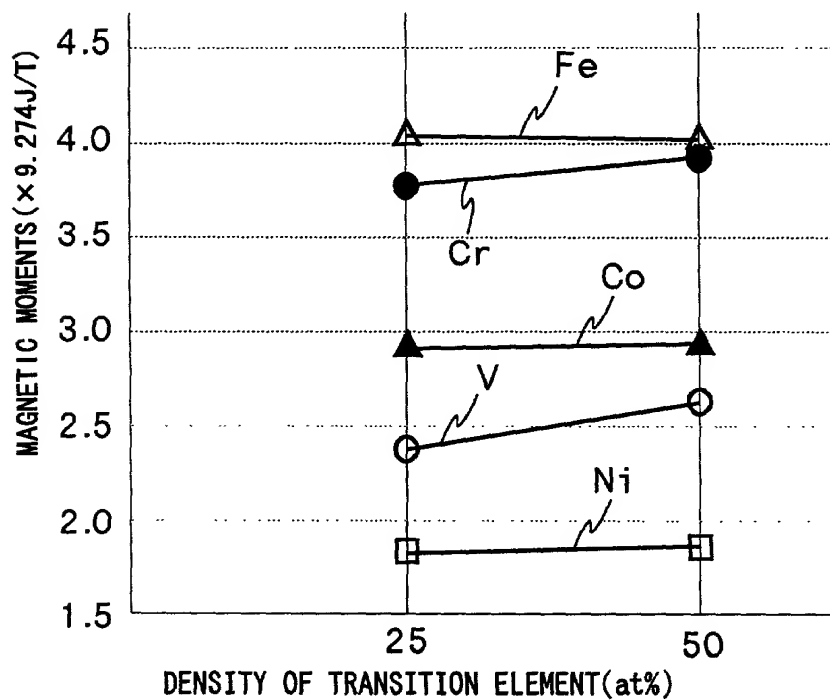


FIG. 4 (a)

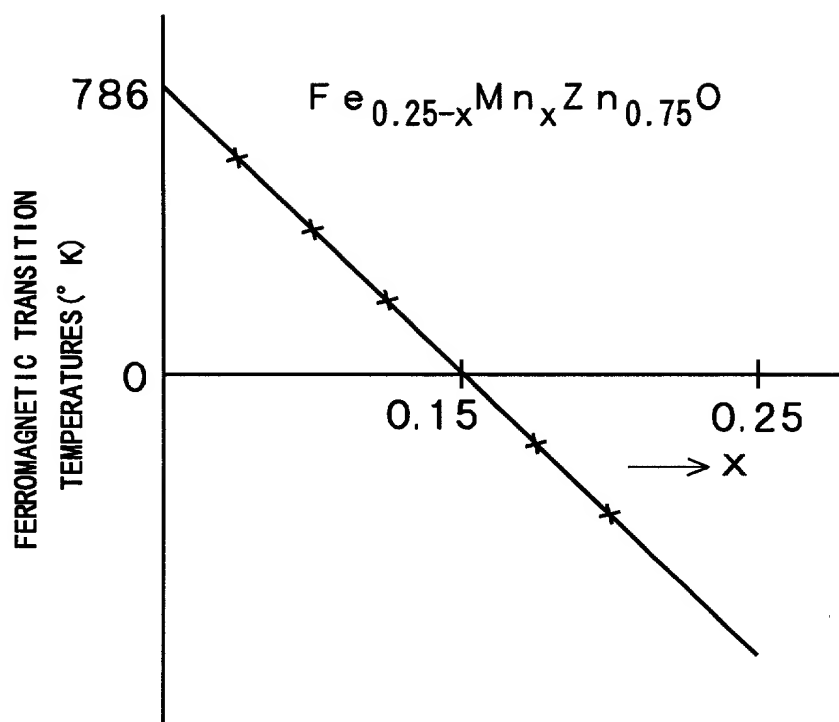


FIG. 4 (b)

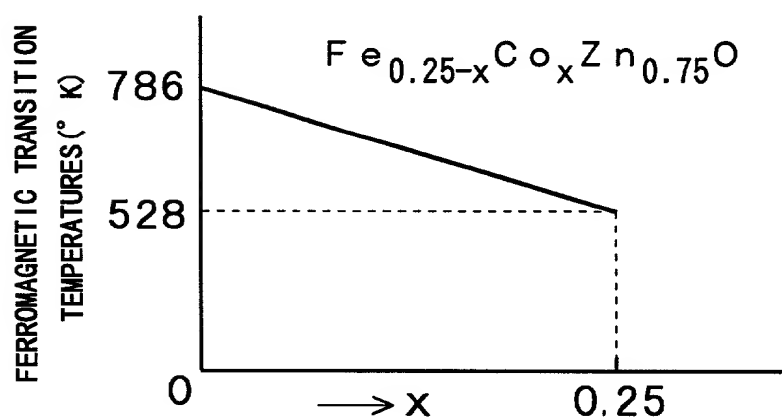
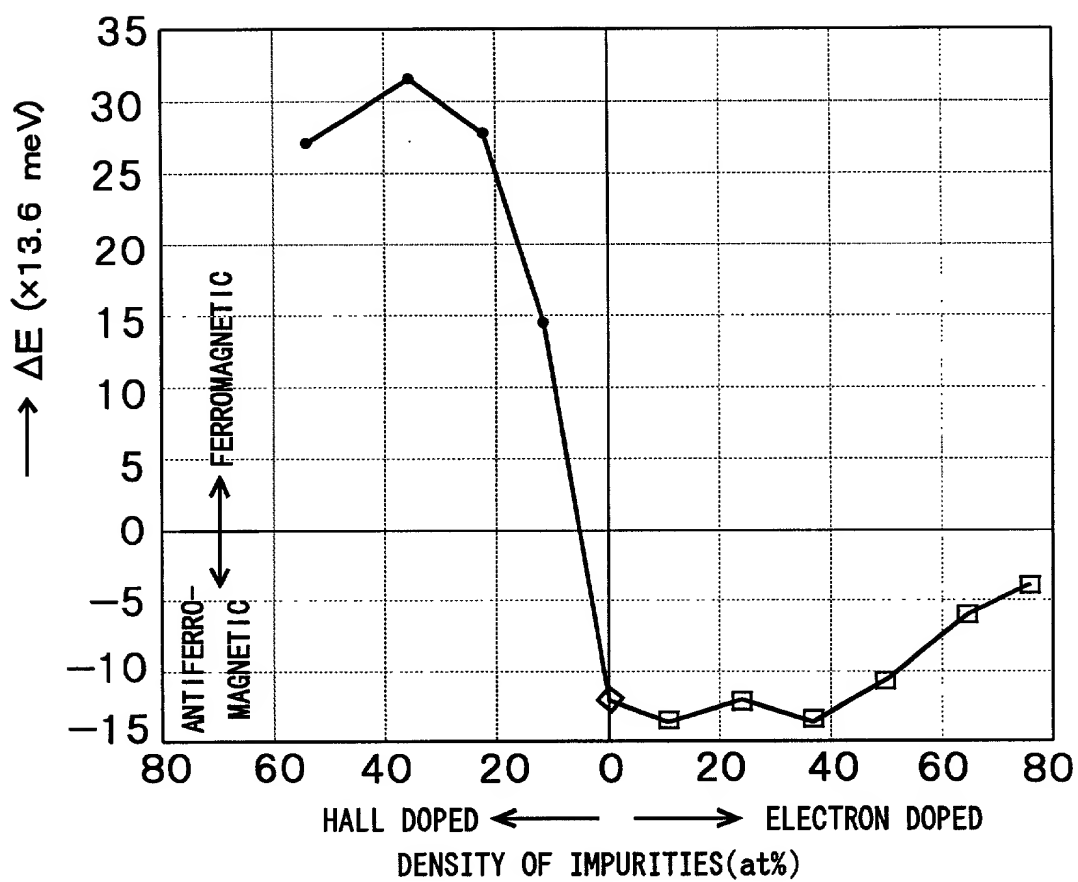


FIG. 5



## Declaration and Power of Attorney For Patent Application

特許出願宣言書及び委任状

## Japanese Language Declaration

## 日本語宣言書

下記の氏名の発明者として、私は以下の通り宣言します。

As a below named inventor, I hereby declare that:

私の住所、私書箱、国籍は下記の私の氏名の後に記載された通りです。

My residence, post office address and citizenship are as stated next to my name.

下記の名称の発明に関して請求範囲に記載され、特許出願している発明内容について、私が最初かつ唯一の発明者（下記の氏名が一つの場合）もしくは最初かつ共同発明者であると（下記の名称が複数の場合）信じています。

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

FERROMAGNETIC ZnO-TYPE COMPOUND INCLUDING

TRANSITION METALLIC ELEMENT AND METHOD FOR

ADJUSTING FERROMAGNETIC CHARACTERISTICS THEREOF

上記発明の明細書（下記の欄でx印がついていない場合は、本表に添付）は、

the specification of which is attached hereto unless the following box is checked:

☐ 月 日に提出され、米国出願番号または特許協定条約国際出願番号を \_\_\_\_\_ とし、  
(該当する場合) \_\_\_\_\_ に訂正されました。☐ was filed on \_\_\_\_\_  
as United States Application Number or  
PCT International Application Number  
\_\_\_\_\_ and was amended on  
\_\_\_\_\_ (if applicable).

私は、特許請求範囲を含む上記訂正後の明細書を検討し、内容を理解していることをここに表明します。

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

私は、連邦規則法典第37編第1条56項に定義されるとおり、特許資格の有無について重要な情報を開示する義務があることを認めます。

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

## Japanese Language Declaration

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私は、米国法典第35編119条(a)-(d)項又は365条(b)項に基づき下記の、米国以外の国の少なくとも一カ国を指定している特許協力条約365(a)項に基づき国際出願、又は外国での特許出願もしくは発明者証の出願についての外国優先権をここに主張するとともに、優先権を主張している、本出願の前に出願された特許または発明者証の外国出願を以下に、枠内をマークすることで、示しています。

## Prior Foreign Application(s)

外国での先行出願

308911/1999

Japan

29/10/1999

(Number)

(Country)

(Day/Month/Year Filed)

(番号)

(国名)

(出願年月日)

(Number)

(Country)

(Day/Month/Year Filed)

(番号)

(国名)

(出願年月日)

Priority Not Claimed

優先権主張なし

☐☐

私は、第35編米国法典119条(e)項に基づいて下記の米国特許出願規定に記載された権利をここに主張いたします。

(Application No.)

(Filing Date)

(出願番号)

(出願日)

(Application No.)

(Filing Date)

(出願番号)

(出願日)

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I hereby claim the benefit under Title 35, United States Code, Section 119(e) of any United States provisional application(s) listed below.

I hereby claim the benefit under Title 35, United States Code, Section 120 of any United States application(s), or 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code Section 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of application.

(Application No.)

(Filing Date)

(出願番号)

(出願日)

(Status: Patented, Pending, Abandoned)

(現況: 特許許可済、係属中、放棄済)

(Application No.)

(Filing Date)

(出願番号)

(出願日)

(Status: Patented, Pending, Abandoned)

(現況: 特許許可済、係属中、放棄済)

私は、私自身の知識に基づいて本宣言書で私が行なう表明が真実であり、かつ私の入手した情報と私の信じることに基づき表明が全て真実であると信じていること、さらに故意になされた虚偽の表明及びそれと同等の行為は米国法典第18編第1001条に基づき、罰金または拘禁、もしくはその両方により処罰されること、そしてそのような故意による虚偽の声明を行なえば、出願した、又は既に許可された特許の有効性が失われることを認識し、よってここに上記のごとく宣誓を致します。

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.



## Japanese Language Declaration

(日本語宣言書)

委任状: 私は下記の発明者として、本出願に関する一切の手続きを米特許商標局に対して遂行する弁理士または代理人として、下記の者を指名いたします。(弁理士、または代理人の氏名及び登録番号を明記のこと)

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith (list name and registration number)

書類送付先

And I hereby appoint as principal attorneys: David T. Nikaido, Reg. No. 22,663; Charles M. Marmelstein, Reg. No. 25,895; George E. Oram, Jr., Reg. No. 27,931; Robert B. Murray, Reg. No. 22,980; Martin S. Postman, Reg. No. 18,570; E. Marcie Emas, Reg. No. 32,131; Douglas H. Goldhush, Reg. No. 33,125; Kevin C. Brown, Reg. No. 32,402; Monica Chin Kitts, Reg. No. 36,105; Richard J. Berman, Reg. No. 39,107; King L. Wong, Reg. No. 37,500; Karen K. Costantino, Reg. No. 35,107; and James A. Poulos, III, Reg. No. 31,714.

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Full name of sole or first inventor

Hiroshi Yoshida

発明者の署名

日付

Inventor's signature

Date

18/10/2000

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第二共同発明者の署名

日付

Second inventor's signature

Date

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Citizenship

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Same as Residence

(第三以降の共同発明者についても同様に記載し、署名をすること)

(Supply similar information and signature for third and subsequent joint inventors.)

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第三共同発明者の署名	日付	Third inventor's signature	Date
住所		Residence	
国籍		Citizenship	
私書箱		Post Office Address	
第四共同発明者名		Full name of fourth joint inventor, if any	
第四共同発明者の署名	日付	Fourth inventor's signature	Date
住所		Residence	
国籍		Citizenship	
私書箱		Post Office Address	
第五共同発明者名		Full name of fifth joint inventor, if any	
第五共同発明者の署名	日付	Fifth inventor's signature	Date
住所		Residence	
国籍		Citizenship	
私書箱		Post Office Address	
第六共同発明者名		Full name of sixth joint inventor, if any	
第六共同発明者の署名	日付	Sixth inventor's signature	Date
住所		Residence	
国籍		Citizenship	
私書箱		Post Office Address	

第七共同発明者名		Full name of seventh joint inventor, if any	
第七共同発明者の署名	日付	Seventh inventor's signature	Date
住所		Residence	
国籍		Citizenship	
私書箱		Post Office Address	
第八共同発明者名		Full name of eighth joint inventor, if any	
第八共同発明者の署名	日付	Eighth inventor's signature	Date
住所		Residence	
国籍		Citizenship	
私書箱		Post Office Address	
第九共同発明者名		Full name of ninth joint inventor, if any	
第九共同発明者の署名	日付	Ninth inventor's signature	Date
住所		Residence	
国籍		Citizenship	
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第十共同発明者名		Full name of tenth joint inventor, if any	
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住所		Residence	
国籍		Citizenship	
私書箱		Post Office Address	

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the application of:

YOSHIDA et al.

Serial Number: New Application

Filed: October 26, 2000

For: FERROMAGNETIC ZnO-TYPE COMPOUND INCLUDING TRANSITION  
METALLIC ELEMENT AND METHOD FOR ADJUSTING  
FERROMAGNETIC CHARACTERISTICS THEREOF

**NOTIFICATION OF CHANGE OF NAME AND ADDRESS**

Commissioner for Patents  
Washington, D.C. 20231

October 26, 2000

Sir:

Kindly change the correspondence name and address for the above-identified  
application to the following:

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Should any fees be due with respect to this paper, please charge Counsel's Deposit  
Account No. 01-2300.

Respectfully submitted,

ARENT FOX KINTNER PLOTKIN & KAHN



By: George E. Oram, Jr.  
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10/30/2000 EFLQRES 00000002 09696013

02 FC:581

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